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With the equipment requested in the associated Dump proposal (DUMP Topic #4, same title as above) and the graduate student support requested herein, we envision several new avenues of research. The tunable, narrow bandwidth output of the injection seeded system will serve as a near-infrared spectroscopic source for imaging combustion products at Wright Patterson Air Force Base (WPAFB). The anticipated energy output (30 mJ, 10 ns pulses) will allow multi-point or line measurements that provide much better diagnostic information than the currently used single point methods. We also envision utilizing the source for atmospheric pollutant sensing in laser radar/differential absorption lidar sensors or in multispectral laser radar target identification systems. Exhaust plumes, even oil pipeline leaks, and targets hidden in foliage can be easily detected by such systems. The pulsed source permits retrieval of range information as well. Finally we suggest some fundamentally new methods of building and tuning wavelength agile sources based on QPM materials structures. One example is a single crystal frequency tripler.

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Wavelength Agile Spectroscopic Sources Based on Quasi-Phase Matched Structures

Abstract

We recently proposed to develop a laser-based, tunable spectroscopic source for tomographic combustion diagnostics. Our source will use a pulsed, high-energy Nd:YAG laser to pump an injection-seeded optical parametric oscillator. Such a configuration gives both high energy (for imaging diagnostics) and narrow bandwidth (for high resolution) that is tunable over the entire near-infrared band. The optical parametric oscillator itself will utilize the innovative technology of quasi-phase matching (QPM) via periodic domain reversal in ferroelectric nonlinear optical crystals. This "engineerable" material system is an enabling technology for many Department of Defense and commercial applications.

With the equipment requested in the associated DURIP proposal (DURIP Topic #4, same title as above) and the graduate student support requested herein, we envision several new avenues of research. The tunable, narrow bandwidth output of the injection seeded system will serve as a near-infrared spectroscopic source for imaging combustion products at Wright Patterson Air Force Base (WPAFB). The anticipated energy output (30 mJ, 10 ns pulses) will allow multi-point or line measurements that provide much better diagnostic information than the currently used single point methods. We also envision utilizing the source for atmospheric pollutant sensing in laser radar/differential absorption lidar sensors or in multispectral laser radar target identification systems. Exhaust plumes, even oil pipeline leaks, and targets hidden in foliage can be easily detected by such systems. The pulsed source permits retrieval of range information as well. Finally we suggest some fundamentally new methods of building and tuning wavelength agile sources based on QPM materials structures. One example is a single crystal frequency tripler.

The new research that this grant will support is a natural extension of our collaboration with WPAFB in the areas of 1) laser spectroscopy as applied to combustion diagnostics and 2) periodically-poled crystals for active laser-based sources. We request funds to support one graduate student for each of these topics and for one additional undergraduate laboratory assistant. Drs. Perry P. Yaney and Vince Dominic will serve as thesis advisors in the respective topics. This effort will provide an excellent opportunity for training in an area that promises to be important to the needs of the Department of Defense. The Department of Physics of the University of Dayton has a well-equipped and newly-renovated optical spectroscopy laboratory (via a NSF University Research Facilities Grant) designed for optical probe measurements in combustion media. The proposed OPO laser system will be developed in this laboratory.

Wavelength Agile Spectroscopic Sources
Based on Quasi-Phase Matched Structures

Optical parametric oscillators (OPO's) have long held great promise as tunable spectroscopic sources. A new method that vastly improves OPO devices utilizes quasi-phase matching (QPM) structures in ferroelectric crystals.[1] Quasi-phase matching improves the device performance by allowing access to the largest nonlinear optical coefficient of a given crystal while simultaneously insuring proper phase matching at virtually any wavelength within the transparency band of the crystal. One can tailor the QPM interaction to suit application needs by controlling the periodicity, location, length, and width of the structure. Curiously, QPM was one of the first phase matching techniques suggested[1] but its use has only recently exploded in popularity.[2-8] In collaboration with Dr. Larry Myers (WL/AAJL) we have developed a facility at WPAFB to periodically pole QPM structures into ferroelectric materials. Since the QPM pattern is determined photolithographically,[6] there is great freedom to imagine, design, and implement new device patterns. One already demonstrated pattern is the multiple grating structure described in ref. 8 that allows rapid tunability of an OPO simply by translating the crystal. We will utilize just such a piece in the tunable spectroscopic source described below.

Our injection seeded, tunable optical parametric oscillator is based on periodically-poled lithium niobate (PPLN). A tunable laser diode with replaceable, narrow band (<100 kHz) modules for different wavelengths will provide the single-mode injection signal. The seed laser will be of modular design so that the use of different modules preserves the wide tunability of the overall system. We will lock the cavity to the longitudinal mode of the diode laser using standard techniques (Pound-Drever-Hall).[9] The high energy output opens new doorways in remote sensing and tomographic spectroscopy. The Plasma Physics group at WPAFB has indicated strong interest in using such tunable, high-energy systems in combustion diagnostics.

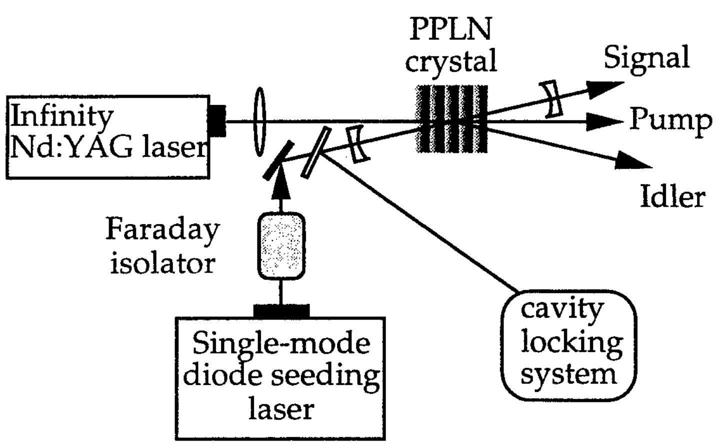

Fig. 1 Schematic view of the injection-seeded, high-energy, tunable source. We design the QPM structure for noncolinear phase matching of the OPO. The crystal is slightly wedged so that there will be no backwards traveling pump, signal, or idler beams. The cavity locking system insures that the signal cavity is locked to the single longitudinal mode of the seed laser by adjusting the position of a piezoelectrically mounted mirror. The seed laser is modular: to switch wavelengths we simply replace the semiconductor laser with one that operates at the desired wavelength. We can seed from 630 nm to 2.2 μm. We pump with 1.064 μm, 532 nm, or 355 nm.

Spectroscopic Use of Injection Seeded OPO

The injection seeded, narrow bandwidth, high energy system will be quite useful for spectroscopic applications. The temperature and/or the concentrations of both major and minor species in combusting gas flows have been characterized by a wide variety of laser-based combustion diagnostics over the years. The optical techniques of choice are Raman spectroscopy, Rayleigh scattering, laser-induced fluorescence (LIF), and coherent anti-Stokes Raman spectroscopy (CARS).[10] One of the main difficulties in using these techniques is the accurate measurement of species concentrations. The traditional method for accurate measurements of species concentration and line strengths of a gas is optical absorption. Recently, the nonlinear technique of degenerate-four-wave-mixing (DFWM) has been developed which provides an absorption measurement at a point.[11] Furthermore, DFWM can be configured

to generate two-dimensional profiles, but with a considerable increase in the pulse energy required. By utilizing DFWM in the infrared region (1-10μm) virtually every minor species can be accessed. At this time, there is no satisfactory tunable laser system capable of providing high energy (tens of millijoules) in this region. The current technology uses low (1-10 mW) cw tunable diode lasers to scan a narrow (10-20 GHz) range with high (tens of MHz) resolution.[12]

Some of the molecular species important to the chemical kinetics of a flame are given in Table 1. Initially, the injection seeded system will be designed to permit detection and measurement of CH_4 (methane) because methane can be provided in a simple laboratory chamber over a wide range of low pressures. These initial studies will be used to evaluate the performance of the OPO laser system. An effort will also be made to compare straight IR detection measurements with measurements using nonlinear up-conversion techniques to move the signals into the wavelength range suitable for photomultiplier detection. Following system checkout, measurements on one or more of the species listed in Table 1 in a simple laboratory methane flame will be carried out. In hydrocarbon flames, formaldehyde is an important intermediate species in combustion chemistry and CH_3 is a precursor to soot formation. NO and NO_2 are hazardous pollutants generated in atmospheric combustion. These particular species will be among the candidates for the proposed flame measurements.

Molecule	Wavelength (μm)	Molecule	Wavelength (μm)
CO	4.7	H_2O	2.7
CO_2	4.3	NO	2.67
CH_3	3.4	NO_2	3.1
CH_4	1.67, 3.3	OH	2.8
CH_2O	3.5		

Table 1 Spectral regions within 1 to 4 μm for selected molecules in gaseous combustion.

The final goal will be to design a DFWM measurement system around the OPO laser system which can provide point measurements of molecular concentrations in a flame. A long-term goal of this final activity will be to devise a scheme which will utilize the line or planar imaging capability of DFWM.[13] The proposed system will provide a significant contribution to laser-based combustion diagnostics by making it possible to obtain concentrations of important intermediate molecular species which are currently difficult or impossible to measure optically with good statistical accuracy in a turbulent combustion medium. The results of these studies will provide new data that will increase the accuracy of the chemical kinetic codes used for combustion modeling and prediction. Moreover, the techniques developed in the proposed program will be applicable to a wide range of optical probe measurements such as in plasmas, atmospheric studies and laser direction and ranging (ladar) applications.

Brief Synopsis of Additional QPM Developments

We give only a brief synopsis of the additional QPM research that the we will pursue:

1) The ability to custom design the wavelength response of QPM crystals opens some very interesting possibilities. One example that we are interested in pursuing is a single-crystal frequency tripler (impossible to do without QPM). Such a device is of great interest for optical information storage, atmospheric remote sensing, and rapid prototyping.

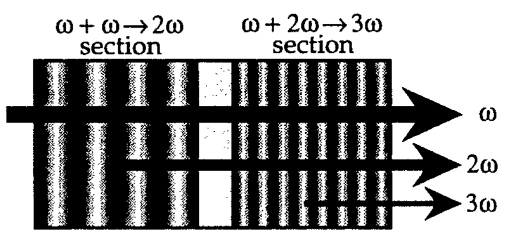

Fig. 2 Spatial control of the QPM structure required for third-harmonic generation. The second-harmonic generation section mixes $\omega+\omega\rightarrow 2\omega$. The 2ω output then mixes with the fundamental to give the third harmonic output. By using two different QPM regions in a single crystal we eliminate the need for two separate mixing crystals. The periodic structure in the $\omega+2\omega\rightarrow 3\omega$ section is much finer than in the frequency doubling section.

2) Achieving the short periodicities required for devices such as that shown above is quite a challenge. We propose investigating the simultaneous use a patterned optical field to assist the standard electric-field domain reversal technique. Such a light-assisted poling regimen will improve the short periodicity devices by utilizing the photoconductivity of the crystal to help control the fringing electric fields during the poling process.

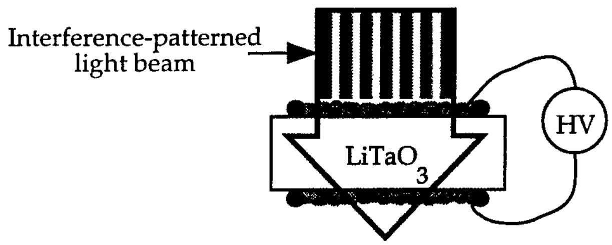

Fig. 3 Opto/electrical poling in $LiTaO_3$. The light beam increases the crystalline conductivity thus helping to reduce the fringing fields leading to better confinement of the domain-reversed region. Notice that we pattern both the applied electric field and the optical field. This is a marriage between the patterned field poling technique and the patterned optical/uniform field poling technique, both of which have been previously attempted.

Summary

The primary thrust of the proposed research is to create a high energy, narrow bandwidth, spectroscopic source by injection seeding an optical parametric oscillator based on quasi-phase matched structures in $LiNbO_3$. The applications of such a system to monitoring previously undetectable species and the improvement over the state-of-the-art in combustion diagnostics by allowing tomographic (area-wide) determinations was presented. We are excited about several unique possibilities for wavelength shifting devices that are afforded by the "engineerability" of QPM structures including: simultaneous two-color OPO's, single-crystal frequency tripling, and switchable Bragg grating devices. While the injection-seeded, high energy spectroscopic source has a declared customer in the Combustion Group at WPAFB, we believe that QPM structures allow so many unique possibilities (freedom of design) that the ancillary research topics discussed here will also breed important innovations for other DOD and commercial customers. The research program discussed herein will produce graduates well trained to join this burgeoning technology race.

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